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Exposure Assessment: A Key Issue in Aquatic Toxicology

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ABSTRACT: Environmental exposure assessment, an intermediate step in risk analysis, ties projected volumes of toxic materials entering the environment to specific environmental effects. Thus, prediction of the environmental fate and distribution of a toxic compound is an integral part of any hierarchical scheme of risk analysis.

Environmental exposure assessment is a new and emerging science, and, although some processes have not been adequately characterized, tests for the most significant ones (volatilization, hydrolysis, oxidation, etc.) are available. Both screening tests and detailed protocols are presently available for determining the process kinetics for selected chemicals exposed to differing environmental conditions. Preliminary exposure assessments can be made from the screening data if the following rate processes are estimated: microbial degradation, chemical transformation, photolysis, hydrolysis, ionization, and partitioning. Because these processes can act either synergistically or competitively, depending upon the chemical structure of the material and the conditions of the environment, great care must be taken to conduct the tests in sequential order.

The greatest unrealized potential for exposure assessment lies in the use of mathematical modeling techniques. When environmental exposure models are coupled with environmental effects models, powerful tools for risk assessment emerge. Such sophisticated models need to be refined and verified, however, before risk analysis becomes accurate and cost-effective.

KEY WORDS: analysis, chemical properties, environment, mathematical models, risk analysis, exposure analysis, toxicology, pollution, aquatic toxicology

The problem of determining the human and environmental risks associated with the manufacture, use, and disposal of chemicals is a major national concern. The current involvement of the various operating and regulatory offices within the U.S. Environmental Protection Agency (EPA) in implementing the Toxic Substances Control Act of 1976 reflects this

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broad interest. It is essential, however, to regard this paper as a report of research concepts and results and *not* as a statement of existing or pending EPA policy.

This paper contains:

1. Discussions of the relative roles of aquatic pollutant loading, exposure analysis, and toxicological effects analysis in conducting environmental and human risk analyses.
2. An approach for conducting exposure analysis.
3. Discussions of the need for developing integrated risk analysis models and their advantages for environmental managers.

The involvement and synthesis of many scientific disciplines in the development of risk assessment techniques requires that terms be clearly defined to prevent semantic disagreements. Thus, in this paper a distinction is made between the terms analysis and assessment. Here, analysis describes the collection and examination of technical and scientific data, whereas assessment includes an evaluation of both the scientific data and the social, economic, and political factors that must be considered in reaching an ultimate decision on the prohibition, control, or management of chemicals in the environment. In this context, risk assessment and hazard assessment are used synonymously.

Environmental Risk Analysis

Following this approach, "risk analysis" encompasses: (1) the systematic collection, integration, and evaluation of environmental loading analysis data; (2) environmental exposure analysis data; (3) human and environmental toxicological effects analysis data (Fig. 1). Thus, in order to conduct a reliable analysis, the toxicological dose-response parameters and data must be compatible with and dependent on the environmental exposure parameters and data. Described another way, environmental exposure analysis is an intermediate step in any risk analysis that ties the volume and distribution of a chemical entering the environment to specific human health or environmental effects. Quantitatively, the pollutant concentration and the duration of exposure are governed by:

- (a) the environmental loading rates;
- (b) the net results of the magnitude and rates of the competing transport, transformation, and degradation (fate) processes acting on the chemical; and
- (c) the physical, chemical, and biological characteristics of the receiving environments.

Environmental Loading Analysis

Chemicals may enter the environment (1) at the time of production

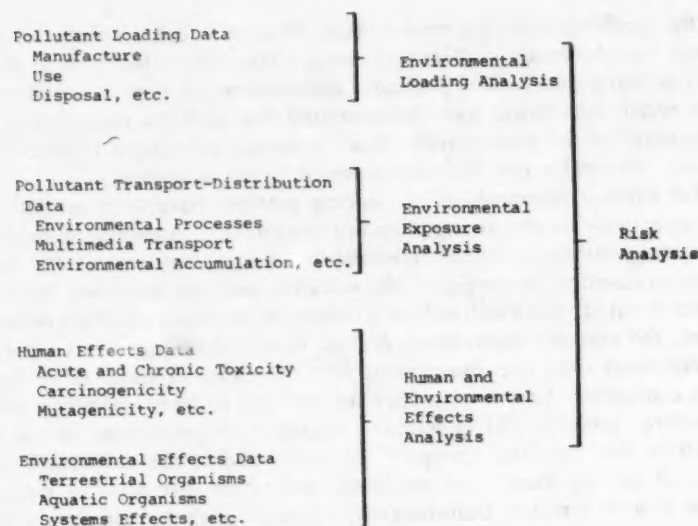


FIG. 1—Environmental pollutant risk analysis.

through direct losses from the production process as solid, liquid, or gaseous waste products are discarded; (2) as the principal manufactured material is packaged, transported, and delivered to intermediate or final users (accidental spills, etc.); or (3) as the product is used or as products containing the materials of concern are used and discarded.

For some materials, precise information may be available on the rates of discharge into various compartments of the environment. For most materials, however, estimates must be made, taking into consideration the projected volumes produced or imported and the anticipated distribution, use, and discard patterns. Regardless of the type of data, the procedures used for loading analysis should integrate and reduce the pollutant inputs to time-series concentrations at specific locations for the aquatic, atmospheric, or terrestrial media. The degree of resolution of the time functions is extremely important and is dependent on both the fate and the effects of the chemical in the environment.

Environmental Exposure Analysis

Once in the environment the distribution, concentration, and structure of a chemical at a given point in time are the net result of a set of basic physical, chemical, and biological processes that transport, degrade, and transform the pollutant either simultaneously or in sequence. The rates at which these processes actually affect a material are functions of both its initial chemical structure and the biotic and abiotic conditions that exist

in the particular receiving environment. Thus, although a number of processes can theoretically affect a chemical's fate, only a few (one to three) may be significant for any particular environment. (It is important to note that recent experiences have demonstrated that both the identity and the magnitude of the predominant "fate" processes affecting a chemical can change drastically from one environmental system to another.)

For existing chemicals whose loading patterns have been established, properly designed monitoring programs can provide accurate environmental exposure estimates. Without minimal knowledge, however, of the basic physical-chemical properties of the materials and the dominant fate processes involved, along with at least a limited characterization of the environment, the exposure information derived from monitoring data cannot be extrapolated from one situation to another, either in space or in time, with confidence. Exposure analyses for both old and new chemicals must, therefore, provide valid quantitative estimates or predictions of the net result of the "fate and transport" processes and of the magnitudes and rates of the significant environmental parameters that drive or regulate these process kinetics. Unfortunately, exposure analysis represents a new and emerging science and the state-of-the-art knowledge is not uniformly distributed across the various environmental media. Moreover, the newly developed procedures and techniques for determining the process kinetics have not been in use long enough to gain general acceptance by the scientific and technical community.

The exposure analysis techniques for pollutants entering freshwater ecosystems appear to be more advanced than those for other media. Sanders [1],² Lassiter et al [2], and Baughman and Lassiter [3], reporting on research at the EPA's Environmental Research Laboratory (ERL) in Athens, Ga., have described a strategy or approach applicable to exposure analysis that includes procedures for characterizing the most significant fate and transport processes that influence organic and inorganic chemicals in freshwater environments and, more importantly, a method for quantitatively analyzing the data. Both laboratory screening tests and detailed analytical procedures have been developed and tested for determining the extent and rates of various processes when selected chemicals are exposed to ranges of conditions representing fluctuations observed in natural environments. These processes are:

ionization	partitioning
hydrolysis	volatilization
photolysis	microbial degradation

Baughman and Lassiter [3] describe and discuss each of these processes

²The italic numbers in brackets refer to the list of references appended to this paper.

in detail, so only a brief summary of their importance will be presented here.

Ionization—Equilibrium relationships, such as the acid-base dissociations, are extremely important in determining the fate and distribution of both organic and inorganic chemicals in aquatic systems. Because the equilibrium relationships govern the concentration of the active chemical species available to participate in the other fate reactions (for example, volatilization, partitioning, etc.), both the rates and the magnitude of the net results can be affected. Ionization is usually rapidly accomplished in completely mixed systems.

Hydrolysis—Many chemicals will hydrolyze once discharged into water. The hydrolysis reactions may follow a number of different mechanisms; however, the kinetics are usually functions of both hydrogen ion concentration (pH) and temperature. Hydrolysis may occur nearly instantaneously or may take several months.

Photolysis—Chemicals can be degraded or transformed in aquatic systems by direct absorption of light and by photosensitized reactions. Because the photolytic rates are functions of the intensity of specific wavelengths of light reaching the chemical, the absorption spectrum of the water, the depth attenuation of the significant wavelengths, the time of day, and the season of the year are all factors that must be considered. Because both direct and indirect photolysis can occur simultaneously, the sum of their rates will equal the net photolytic rate. This net rate is only slightly affected by temperature or ionic strength, and the degradation or transformation period may range from hours to years.

Partitioning—Baughman and Lassiter [3] state that the partitioning of chemicals between the water, the plants and animals, and the abiotic solids is probably the most significant fate and transport process in aquatic systems. The partitioning of a chemical in a natural system is a complex process that occurs by several different mechanisms that depend upon the characteristics of the chemical and the nature of the sorbents present in the water. Partition coefficients for both sediments and microorganisms have been found to be relatively consistent (with agreement within a factor of 2 or 3) from one location to another and between various microorganisms. Adjustment for the effects of the organic content in sediments reduces the variability even more.

Like ionization, partitioning affects the net fate of a chemical by effectively reducing the concentration of the chemical in solution. This, in turn, affects the rates of the other competing processes. Although partitioning is a relatively rapid process, the reactions are reversible in most instances. It is also important to note that for some classes of chemicals the strongest sorption is of the fine suspended particles so that, even though sorbed, the hydrodynamic movement of those materials is similar to that of chemicals in solution.

Volatilization—The relative importance of the volatilization process in determining the concentration of chemicals in aquatic systems has been demonstrated many times; however, methods for predicting the process rates are new. Like the exchange of oxygen and other gases at the air-water interface, molecules of the chemical must be moved to the surface of a water body, where the mass transfer takes place. Thus, the net volatilization rate is a function of the mixing or turbulence within the system, as well as of the basic properties of the chemical (Henry's law constant) and the concentration of the chemical in solution. Thermal stratification, wind and wave actions, and other physical factors can drastically affect the net volatilization.

Microbial Degradation—Recent laboratory and field studies indicate that the relative significance of microbial degradation processes may be less than previously believed. The determination of the relative significance of microbial degradation processes in aquatic systems is extremely complex, and state-of-the-art procedures can only provide gross estimates at this time. The rates and extent of these processes depend upon a number of interacting factors, including the:

- (a) availability of enzyme systems for degrading particular chemicals;
- (b) presence of sufficient numbers of indigenous organisms to sustain the process;
- (c) concentrations of the target chemical, plus concentrations of other major and minor nutrients required for growth;
- (d) absence of toxic or inhibiting chemicals; and
- (e) availability of favorable growing conditions (temperature, pH, etc.).

Thus, estimates based on kinetic data obtained in the laboratory, where growth conditions may be more favorable, should be higher than the process rates actually observed in nature. The proper analysis of laboratory data, however, can place the relative process rates in perspective with other competing physical and chemical processes.

Other significant transport and transformation processes will be added to the evaluation protocols as their mechanisms are characterized and their rates and extent, under various environmental conditions, are quantified.

To date, some or all of the significant fate processes for approximately 44 different organic chemicals (Table 1) have been studied and characterized [4-23], using the procedures developed or adopted at the Athens ERL. In order to verify that other laboratories could conduct similar analyses, the Stanford Research Institute (SRI), under EPA contract, characterized an additional 13 chemicals (Table 2), using the generalized protocols [24].

Evaluative Model

Basic to the exposure analysis procedure for aquatic environments is the development and incorporation of a data analysis system called an "evalua-

TABLE 1—Significant fate processes for selected organic chemicals identified by the Athens ERL.

Chemical Studied	Process Studied				Partitioning (biota or sediment)
	Micro-bial Degrada- tion	Chemical Transforma- tion	Photo- chemical Transforma- tion	Vola- tiliza- tion	
Atrazine	X	X	X		
Captan	X	X	X		
Carbaryl	X	X	X		
2,4-D esters	X	X	X	X	X
Diazinon	X	X	X		
Malathion	X	X	X		
Methoxychlor	X	X	X		X
Parathion	X	X	X		
Toxaphene	X	X	X		
PCB	X			X	X
<i>m</i> -Chloroaniline	X	X			
3,4-Dichloroaniline			X		
Aniline	X				
Fenthion		X	X		
Hexachlorocyclo- pentadiene		X			
Dimethylmercury		X	X	X	
DDE		X	X	X	X
DDT		X	X		X
DMDE		X	X		
Propham	X	X	X		
Trifluralin			X		
Chlorpropham	X	X	X		
Methylmercuric chloride		X	X		
Diphenylmercury		X	X		
Phenylmercuric acetate		X	X		
Triethylphosphorothioate			X		
2,5-Dimethylfuran			X		
Dibenzodioxin			X		
Dibenzofuran			X		
2,4,6,2',4',6'-Hexachloro- biphenyl	X				X
2,4,2',4'-Tetrachlorobiphenyl	X				
2,5,3',4'-Tetrachlorobiphenyl	X				X
Cyanazine	X				
Diphenamid	X				
Paraquat	X				X
Pyrene					X
Naphthalene					X
Phenanthrene					X
Tetracene			X		X
Anthracene			X		X
9-Methylanthracene			X		X
2-Methylnaphthalene					X
1-Methylnaphthalene					X
Vinyl chloride	X	X	X	X	

TABLE 2—Significant fate processes for selected organic chemicals identified by the Stanford Research Institute.

Chemical Studied	Process Studied				
	Micro-bial Degrada- tion	Chem- ical Transfor- mation	Photo- chemical Transfor- mation	Vola- tiliza- tion	Parti- tioning (biota or sedi- ment)
3,3'-Dichlorobenzidine	X		X		X
Lindane	X				
p-Cresol	X	X	X	X	X
Dibenzothiophene	X	X	X	X	X
Methyl parathion	X	X	X	X	X
Mirex	X	X	X	X	X
Carbazole	X	X	X	X	X
Benzolapyrene	X	X	X	X	X
Quinoline	X	X	X	X	X
Benzo (f) quinoline	X	X	X	X	X
Dibenzo (c,g) carbazole		X	X	X	X
Benz (a) anthracene	X	X	X	X	X
Benzothiophene	X	X	X	X	X

tive" model. Everyone defines models differently; in this context, an evaluative model is a mathematical representation of reality, used to provide a quantitative estimate of the net result of a number of competing and interacting transport and transformation processes subjected to a broad range of environmental conditions. Simply stated, an evaluative model is a quantitative decision process expressed in mathematical language.

In developing the concept of evaluative models for exposure analysis, the Athens ERL staff proposed that the environmental characteristics of three physically different aquatic environments (fast-flowing rivers, ponds, and deep stratified lakes) be modeled to simulate representative ecosystems, as opposed to specified natural systems. It was anticipated that in characterizing these representative ecosystems for the models, quantitative descriptions developed by the leading experts in each area could be combined and utilized. The laboratory-derived process kinetics data could then be fed into the model along with projected loading rates, with the model outputs providing the relative concentrations and distribution patterns for any test chemical within the three systems. In short, specific data on a specific chemical would be evaluated in terms of representative environments.

The evaluative model concept was first tested using vinyl chloride as a subject chemical. Hill [25], reporting on the results, concluded that the most significant transport process was volatilization. Because the rate of exchange of vinyl chloride at the air-water interface was about twice that of oxygen, the chemical was predicted not to persist in water even under worst-case analyses.

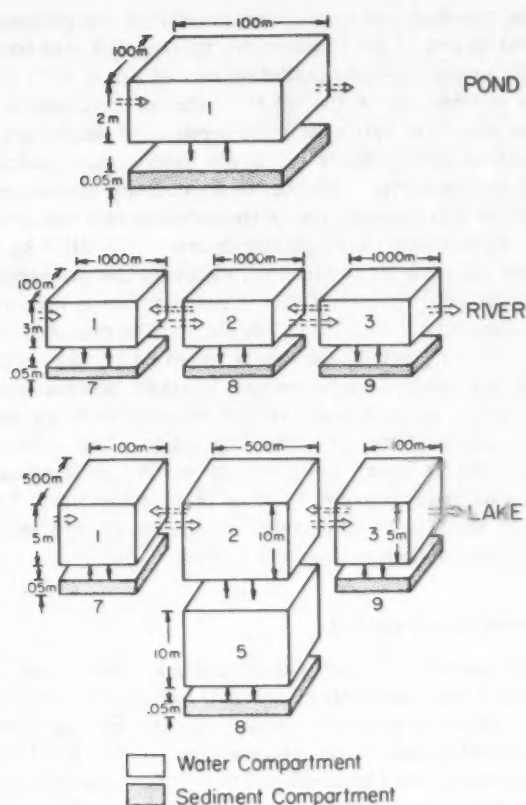


FIG. 2—Physical configurations of a simplified nine-compartment model for ponds, rivers, and lakes.

This concept was further developed at the Athens ERL and through the SRI contract [24], where the initial efforts resulted in the development of a simplified nine-compartment model (see Fig. 2). The input conditions could be changed so that the same model construct would represent the stream, pond, or lake environment. As the contract progressed the model was improved and can now represent up to 99 compartments. More importantly, the fate and transport process kinetics data developed for the 13 organic materials were analyzed using this model, and their relative concentrations and distributions were predicted for the representative stream, pond, and lake systems.

It is also important to note that these model results indicate that individual fate process kinetics data can be very misleading if viewed in isolation from the other competing processes or from the receiving environment.

For example, the disappearance quantum yield for the photolysis of methyl parathion (MP), one of the 13 chemicals investigated, was found to be 1.7×10^{-4} with an average biodegradation rate of 2.5×10^{-7} ml cell⁻¹ h⁻¹. However, a comparison of the relative rates of degradation of MP discharged into a shallow eutrophic pond versus that discharged into a deep oligotrophic lake clearly showed that the predominate pathways and the net rate of disappearance shift between receiving environments. In the eutrophic pond environment, the biodegradation rate was projected to be 1.8×10^{-2} kg MP/h, and the photolysis rate 4.8×10^{-6} kg MP/h. Conversely, using the same basic rate coefficients for the discharge of MP into the oligotrophic lake environment, the biodegradation rate was predicted to be approximately 5.2×10^{-5} kg MP/h, and the photolysis rate approximately 7×10^{-2} kg MP/h. Obviously, it would be almost impossible to place significant maximum or minimum limits on process rates if they are isolated from the characteristics of the various receiving environments.

Evaluative models have yet to be applied to whole environments, for which intermedia transport and distribution may be significant in determining the combined exposure to man or to ecosystems. The concepts, however, have been shown to be valid for aquatic systems, and research is underway to develop such multimedia decision tools.

Environmental Effects Analysis

To provide deciding officials with functional "risk analysis" techniques, the human and environmental effects data should be compatible with the loading and exposure evaluative model outputs. For maximum compatibility, individual-organism or whole-system toxicological dose-response model subroutines should be developed to interface directly into the analysis package. If this is not possible, it would still be beneficial to know the:

- (a) modes and rates of uptake and elimination,
- (b) saturation levels (half-saturation constants) attained,
- (c) time varying dose responses, and
- (d) effects of the physical-chemical environment (temperature, pH, water hardness, etc.) on the above parameters.

Discussion

Today in the environmental field, when the term model is used, there is an immediate response of either openness or complete rejection of the ideas being conveyed. Unfortunately, rejection is often caused by a lack of understanding of the basic concepts of model development and use or by some past unpleasant or unrewarding experience associated with improper application of modeling technology.

The scientific community (for example, Monod [26]) has long utilized

models to express observations and hypotheses. However, the use of mathematical models in decision making has gained little acceptance, although every person develops and utilizes a form of model for each decision that is made. The model may be only in the mind, yet it involves: (1) assembling and assessing pertinent information, (2) predicting the outcome of alternatives, and (3) weighing the alternatives and selecting the "best" solution. Thus, a successful individual is one whose decision modeling provides acceptable solutions more often than not, and the true expert is one whose record demonstrates a high degree of success.

Unfortunately, for those required to conduct human and environmental risk analyses, the chemical data bases are so large, and there are so many interacting factors, that individuals can seldom track the decision process to a successful conclusion without the aid of more sophisticated tools. Thus, it should be a logical and necessary progression for environmental managers to advance from the simple mental decision process to more complex decision models that utilize the best state-of-the-art tools and techniques available.

There are several major advantages that can be derived from using risk analysis models for decision making. Models provide:

1. Better problem definition. In the process of developing or selecting models for risk analysis, the decision maker is required to analyze carefully the problem at hand and to list all the major contributing factors that are known. This analysis focuses on interactions and relationships that might exist and reduces the chance of omitting pertinent information.
2. A guide for data collection. When a model is developed during the initial phase of a problem analysis it serves as a basis for optimum data collection. Because the data are used to calibrate the model, there is usually a large saving in time and facilities, and the persistent problem of collecting inappropriate or unneeded data is minimized.
3. A framework for assessing data. A well-designed model automatically provides the format for data analysis. Just as the form of the model dictates what data are required, models can also provide automated procedures for extracting rate coefficients, saturation or limiting values, and other information from raw data.
4. An ability to handle complex interactions. Properly developed decision models permit many competing factors that occur at different times and at different rates to be evaluated simultaneously, so that the net results can be estimated.
5. An ability to predict. Rational decision models that couple mathematical descriptions of the various interacting factors provide the capability of predicting responses to combinations of conditions that fall outside of the range of the historical data used for the initial model calibration. Probably the greatest benefit to be derived by decision makers from using risk analysis models results from the ability to test any number of hypo-

thetical situations by varying the combinations of input conditions through projected extreme values. These extreme conditions may represent actual measured values or they may be generated by mathematical and statistical techniques (that is, stochastic methods). The resulting range of model output values can readily be used in selecting optimum solutions from the alternatives proposed.

6. The possibility of detailed examination of the decision processes. Because the development of risk analysis models requires that all of the assumptions, data bases, and outputs be recorded, the entire decision process can be examined by all interested parties. This factor can be of great assistance in presenting and explaining management decisions to environmental, industrial, and citizen's groups, whose support is vital to the judicious management of environmental chemicals.

It is essential to point out that models are only "partial truths" because our ability to "represent reality" is limited by our inability to perceive, measure, or mathematically describe real environments, systems, organisms, things, or events with total accuracy. In spite of this lack of knowledge, however, the step-by-step, systematic approach required for the development of mathematical systems models offers the most rational method of making management decisions relative to the complex issues of chemical controls.

Conclusion

An exposure analysis system has been proposed that combines laboratory-derived fate and transport process kinetic data with quantitative model descriptions of representative environments in order to estimate the net exposures of chemicals to man and to the environment. This modeling procedure has the advantage of providing a routine systematic analysis, incorporating the knowledge of leading experts into the decision process, which can be followed and evaluated on a step-by-step basis. Environmental exposure models can be combined with environmental loading and effects models to provide risk analysis systems techniques for environmental decision officials.

Acknowledgments

The concepts presented in this paper are outgrowths of research by several scientists at the Environmental Research Laboratory, U.S. Environmental Research Agency, Athens, Ga. Their work is gratefully acknowledged.

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